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# Comparison of Pesticide Root Zone Model Predictions with Observed Concentrations for the Tobacco Pesticide Metalaxyl in Unsaturated Zone Soils

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16. ABSTRACT

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# COMPARISON OF PESTICIDE ROOT ZONE MODEL PREDICTIONS WITH OBSERVED CONCENTRATIONS FOR THE TOBACCO PESTICIDE METALAXYL IN UNSATURATED ZONE SOILS

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**Abstract**—The pesticide root zone model (PRZM) was developed to evaluate pesticide leaching threats to ground water for different crops under varying climatic conditions, soil characteristics and cropping practices. In this study, PRZM was evaluated to establish its predictive capability by comparing observed metalaxyl concentration profiles with predicted concentration profiles using field-averaged pesticide data and best estimates for several hydrologic characteristics and pesticide transport properties at field sites in Florida and Maryland. Coefficients of determination for the Florida site at 26, 55 and 85 d after application of 0.33, 0.90 and 0.95, respectively, were obtained when field observations were regressed against model predictions. An overall coefficient of determination for the Maryland site of 0.75 was determined for 14 samplings after metalaxyl application.

Pesticide transformation in the Florida soil was best described by a first-order model. A first-order degradation rate in soil of 0.014 $d^{-1}$ was estimated. Pesticide transformation in the Maryland soil was best described by a phased first-order model. A phase determination point that occurred 30 d after metalaxyl application was used to estimate soil degradation rates. Differences were attributed to the timing of rainfall events and soil infiltration characteristics. Estimated phased first-order degradation rates in soil for the Maryland site were 0.046 and 0.0045 $d^{-1}$ before and after the phase determination point.

Evaluation of PRZM, using best estimates for transport and transformation properties of metalaxyl and limited calibration for water balance, suggests that the model effectively simulates the important processes operating on the pesticide.

**Keywords**—Pesticides Models Performance testing Metalaxyl

## INTRODUCTION

Ground water contamination from pesticide use [1] has received considerable attention during the last few years. This increased attention is due in part to the current production of pesticides that are more water-soluble (and hence more leachable) than earlier formulations [2]. The potentially widespread ground water contamination by agricultural chemicals makes remedial actions difficult because there is no single plume originating from a point source that can be isolated and controlled.

The discovery of pesticides in ground water has led to intensive environmental risk assessments for existing or new chemicals. Mathematical models are increasingly being used to evaluate the interaction of agricultural soils with chemical processes and reactions. Models simulate many of the important hydrologic and chemical processes and reactions occurring in the agricultural field and transform numerical input into a quantitative representation of pesticide movement. Modeling provides a logical, meaningful mechanism to integrate plant and soil properties and chemical processes and reactions so that a pesticide's potential effect on ground water quality can be evaluated.

An important part of model development is its successful demonstration in a carefully designed and comprehensive testing program. Tests of model performance using laboratory data are indicative of the accuracy that can be expected when the model is operated in a screening mode, but they are not sufficient for field-scale predictions. Tests of model performance using site-specific field data more clearly demonstrate the

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predictive capability of the model when site-specific variations such as soil properties are accounted for. The extent to which either kind of test can be performed is limited by the available data and the experimental design of the field experiment through which those data were obtained. Soil system properties that may affect pesticide leaching vary spatially in agricultural fields [3]. The variations associated with these soil properties and/or pesticides generally are not provided in existing databases on pesticide leaching.

Ground water contamination from agricultural pesticides is largely a field-scale problem [4]. Although soil system properties (i.e., hydraulic conductivity) vary from profile to profile in the same field, the modeling goal is not to predict the solute concentration at each point; rather, the macroscopic representation of the entire field is of interest [5]. The evaluation of model performance under "field-averaged" conditions may be useful for indicating a model's predictive capability and utility as a screening tool in evaluating new or existing pesticides. The existence of skewed, non-normal distributions will influence the expected value for field-averaged leaching and must be acknowledged and accommodated when possible.

Site-specific monitoring data can be used to evaluate model performance by comparing model predictions with the site-specific observations. The site-specific information can further be used to determine estimates of pesticide leaching to ground water by "calibrating" the model to the site-specific observations, and subjecting the data to many different hydrologic responses using long-term climatic records. The calibrated model can also be used to help determine what additional research is required to measure pesticides and ground water contamination with sensitivity analyses.

The pesticide root zone model (PRZM) was developed to evaluate pesticide leaching threats to ground water for different crops under varying climatic conditions, soil characteristics and cropping practices [2,4]. PRZM has been applied in a screening mode to simulate the mass of pesticide leached below the plant root zone in corn, sorghum and cotton growing areas [6]. This screening method provides a measure of the frequency and severity of pesticide leaching and/or indicates the necessity for more detailed analyses.

In earlier studies [7,8], PRZM was tested using field-averaged data for the pesticide aldicarb in citrus and potato applications. This article describes performance testing of the model using field data collected for the pesticide metalaxyl (*N*-(2,6-dimethylphenyl)-*N*-(methoxyacetyl)-alanine methyl ester) from sites located in tobacco-growing areas of Florida and Maryland. The physical-chemical properties of water solubility, vapor pressure, molecular weight, melting point and specific gravity have been reported to be $2.54 \times 10^{-2}$ mol $L^{-1}$, $2.2 \times 10^{-6}$ torr (20°C), 279.34, 71.0°C and 1.21, respectively (personal communication, D. Sumner). The data collected for metalaxyl are part of the Ciba-Geigy research and development program on the fate and transport of agricultural pesticides.

## TESTING PROCEDURE

### *Hydrologic characteristics and model calibration*

The sites for the Ciba-Geigy field monitoring program in Florida and Maryland were selected because of the availability of general hydrogeologic information [9–13], their location in major tobacco-growing areas of each respective state [9,14] and landowner cooperation. A comprehensive soils database was used to obtain general soil system properties for each site [15]. These general information sources were used to establish PRZM parameters for hydrologic, soils, crop and runoff-infiltration characteristics [4]. The Florida site (a 3.5-ha field) and the Maryland site (a 0.6-ha field) were planted with tobacco in 1980. The estimated characteristics of each field site are summarized in Tables 1 and 2. Several PRZM-required hydrologic measurements, such as daily pan evaporation and precipitation, were not made at either field site but were estimated from historical data and from monthly precipitation totals.

A representative water balance was estimated using the monthly precipitation totals [16]. The

Table 1. Summary of hydrologic characteristics[a]

| Characteristic[b] | Site | |
|---|---|---|
| | Florida | Maryland |
| Precipitation (cm) | 110.0 | 100.0 |
| Crop root zone (cm) | 45.0 | 45.0 |
| Hydrologic soil group | A | C |
| Soil Conservation Service curve number | | |
| Fallow | 78 | 91 |
| Cropping | 67 | 85 |
| Residue | 72 | 88 |

[a]Data from refs. 4 and 9–15.
[b]Average annual values.

Table 2. Summary of soil characteristics[a]

| Characteristic[b] | Site | |
|---|---|---|
| | Florida | Maryland |
| Soil Conservation Service soil classification | Blanton fine sand | Marlton fine sandy loam |
| Soil texture | | |
| Surface | Sand (0–23 cm) | Sandy loam (0–25 cm) |
| Subsurface | Sand (23–148 cm) | Sandy clay (25–140 cm) |
| Substratum | Sandy clay loam (148–216 cm) | Sandy clay loam (140–152 cm) |
| Field capacity and wilting point potentials | | |
| Surface ($cm^3\ cm^{-3}$) | 0.091, 0.033 | 0.207, 0.095 |
| Subsurface ($cm^3\ cm^{-3}$) | 0.091, 0.033 | 0.339, 0.239 |
| Substratum ($cm^3\ cm^{-3}$) | 0.257, 0.148 | 0.257, 0.148 |
| Percent organic matter | | |
| Surface | 1.5 | 2.5 |
| Subsurface | 1.0 | 1.0 |
| Substratum | 0.5 | 0.5 |
| Soil bulk density | | |
| Surface ($g\ cm^{-3}$) | 1.45 | 1.25 |
| Subsurface ($g\ cm^{-3}$) | 1.55 | 1.30 |
| Substratum ($g\ cm^{-3}$) | 1.65 | 1.30 |

[a]Data from refs. 4, 10, 11 and 15.
[b]Average values.

estimated water balance and hydrologic information provided in Table 1 were used to calibrate PRZM [4] using historical climatic data (1956 to 1984) from nearby meteorological stations in Lake City, Florida, and Beltsville, Maryland. Calibration is an interactive procedure of parameter evaluation and refinement that is required for parameters that cannot be deterministically empirically evaluated for a given site. A site-specific parameter used in the calibration exercise with PRZM was the pan evaporation factor (evapotranspiration). The simulated hydrologic summary for each site is provided in Table 3. The hydrologic summaries indicate a potential differences in the runoff-infiltration-recharge potential between the two sites.

Table 3. Hydrologic summary for Florida and Maryland simulations

| | Site | |
|---|---|---|
| | Florida | Maryland |
| Runoff (cm) | 10.0 | 25.0 |
| Evapotranspiration (cm) | 70.0 | 60.0 |
| Recharge (cm) past crop root zone | 30.0 | 15.0 |

*Pesticide application and field monitoring*

The 3.5-ha Florida field site was treated with 0.5 kg $ha^{-1}$ metalaxyl, applied to the soil surface with a boom sprayer as Ridomil 2E formulated product on 26 March 1980. Uniform applications (with coefficients of variation of less than 20%) have been reported for applications made with boom sprayers where the soil is not disturbed after application (personal communication, C.N. Smith). Tobacco was transplanted to the field 3 d later. The 0.5 kg $ha^{-1}$ application was found to be analytically inadequate for monitoring metalaxyl movement, so a subsequent application of 2.2 kg $ha^{-1}$ was applied to a 1.5-ha subplot using a backpack sprayer (because the tobacco had already been transplanted) on 10 April 1980. Soil core samples from the subplot were collected with a 10-cm bucket auger to a depth of 30.0 cm at 15-cm increments and to a depth of 90.0 cm at 10-cm increments on the day of application (10 April). Subsequent samples obtained on days 26, 55, 85 and 154 were taken to a depth of 90.0 cm at 15-cm increments. Two soil cores, one from the northern half and one from the southern half, of the subplot were taken at each sampling interval. These soil cores were combined to obtain field-averaged metalaxyl residue values.

The 0.6-ha Maryland field site was treated with 2.2 kg $ha^{-1}$ metalaxyl, applied to the soil surface

with a backpack sprayer as Ridomil 2E formulated product on 29 May 1980. Subsequent residue determination did not confirm the 2.2 kg $ha^{-1}$ treatment (an application rate of 0.1 kg $ha^{-1}$ was identified). A second application was made on 9 July 1980. Tobacco was transplanted to the field on 9 July 1980. Soil core samples were collected with a 10-cm bucket auger to a depth of 90.0 cm at 15-cm increments on the day of application (9 July) and on days 15, 30, 44, 49, 61, 76, 91, 106, 121, 135, 152, 219 (13 February 1981), 261 and 287. Four soil cores, one each from the four quarters of the field, were taken at each sampling. These soil cores were combined to obtain field-averaged metalaxyl residue values.

*Pesticide characteristics and model parameter development*

Several metalaxyl transport and transformation properties are required for input into PRZM. The metalaxyl transport property required for PRZM is the distribution coefficient ($K_d$) for soil and water. An organic matter distribution coefficient ($K_{om}$) of 40.0 for Lakeland sand has been reported (personal communication, D. Sumner). (A standard deviation was not provided.) The Lakeland sand $K_{om}$ was used to estimate the distribution coefficient $K_d$ by normalizing [16] for the organic matter content in each soil horizon for the Blanton and Marlton soil series at each field site. A summary of these estimated $K_d$ values is provided in Table 4.

The metalaxyl transformation property required for PRZM is the apparent first-order soil degradation rate constant ($k_s$). Metalaxyl residues were not found deeper than 90.0 cm at either field site. Thus, a first-order soil degradation rate constant was estimated by summing calculated average metalaxyl soil residues in each sampling increment (10 or 15 cm) for the 90.0-cm soil core. This procedure is described by the equation:

$$AR_t = \sum_{i=1}^{N} \frac{T_i R_i}{TD} \quad (1)$$

where $AR_t$ is the average residue in soil core ($\mu g\ kg^{-1}$), $T_i$ is the depth of soil sampling layer (cm), $R_i$ is the average residue in sampling layer ($\mu g\ kg^{-1}$), $TD$ is the total depth of soil core (cm) and $N$ is the number of depth increments. (The average soil residues estimated with Equation 1 assume that detectable (greater than 0.05 ppm) metalyaxyl residues do not exist below $TD$.) A first-order model was used to describe the data. The estimated soil degradation rate constants for the Florida and Maryland sites are provided in Figures 1 and 2.

Table 4. Estimated distribution coefficients ($K_d$) for metalaxyl

| | Site | |
|---|---|---|
| Soil layer | Florida | Maryland |
| Surface | 0.60 | 1.20 |
| Subsurface | 0.40 | 0.40 |
| Substratum | 0.20 | 0.20 |

Data for the Florida site could be described by a first-order model. The data for the Maryland site, however, suggested that the soil degradation rate was not first-order. The apparent non-first-order soil degradation rate of metalaxyl for the Maryland site presents a problem in developing input for PRZM, which requires a first-order degradation rate.

The differences for soil degradation observed between the Florida and Maryland sites could be attributed to differences in the timing of rainfall events and soil infiltration factors. A total of 12.0 cm of precipitation was measured at the Florida site within 26 d of application, as compared with 6.0 cm during the first 30 d at the Maryland site. In fact, only 6.0 cm of rain fell in the 3 months after the first 30 d; precipitation in Maryland in 1980 was only 65% of the yearly average. Because of the drier conditions shortly after application, degradative processes at the soil surface could have proceeded for a longer interval at the Maryland site. The pesticide could have infiltrated into the subsurface at a faster rate in the Hydrologic Group A Blanton soil series of the Florida site. The potential infiltration rate [4] for Hydrologic Group A soils is higher than for the Hydrologic Group C Marlton soils [4] of the Maryland site.

Several investigations have suggested that degradation can proceed at a phased first-order rate for pesticides applied to the soil surface, with the phases determined by the timing of the initial precipitation events [17–19]. These varying phased dissipation rates are thought to be due to different processes (such as volatilization or photolysis) affecting the pesticide's disappearance on the soil surface that do not occur after the pesticide has infiltrated into the soil. A phased first-order model was found to describe the Maryland data when a phase determination point of 30 d post-metalaxyl application was used.

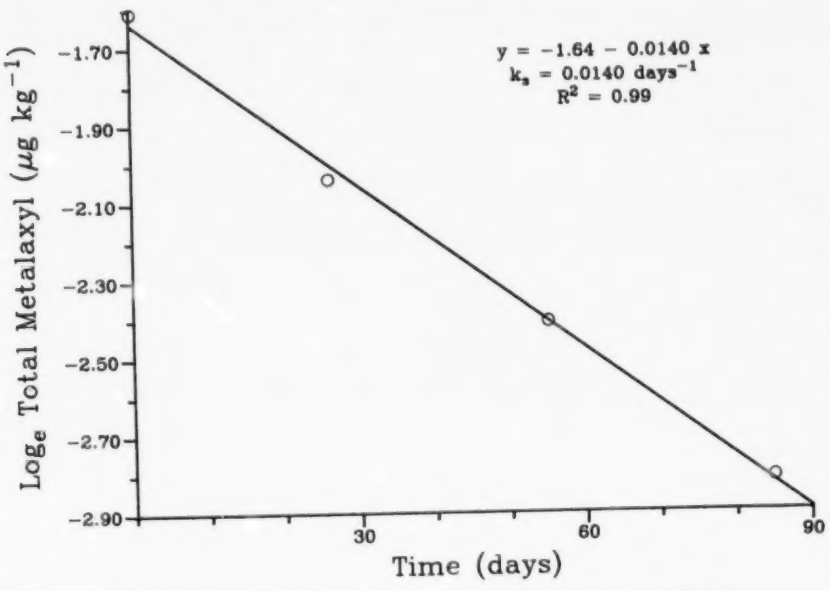


Fig. 1. Estimation of metalaxyl first-order degradation rate in Florida soil.

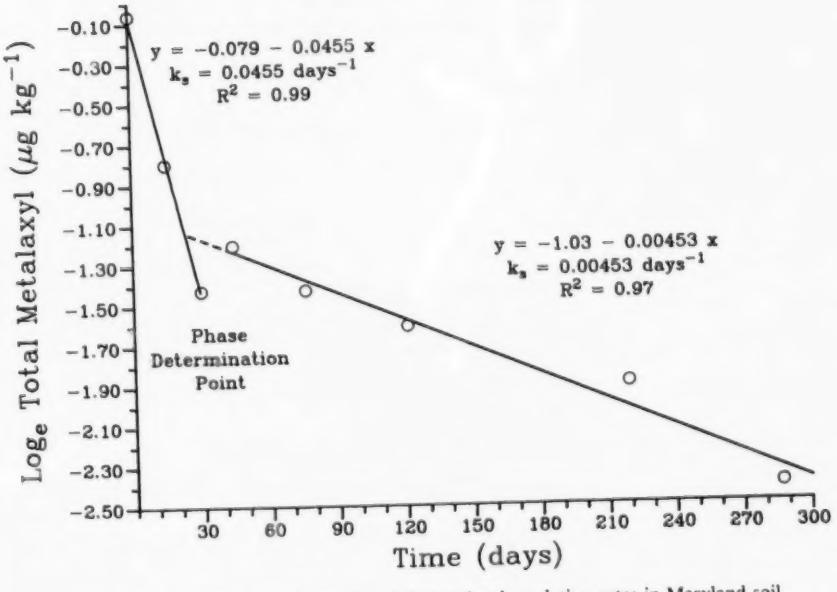


Fig. 2. Estimation of metalaxyl phased first-order degradation rates in Maryland soil.

To evaluate the Maryland metalaxyl monitoring data with PRZM, a two-step procedure was developed. The first step consisted of running the model for 30 d after metalaxyl application using the phase 1 soil degradation rate constant. An initial boundary condition with the mass of metalaxyl calculated in the soil profile from step 1 was established. Step 2 consisted of running the model from

30 d after metalaxyl application using the phase 2 soil degradation rate constant.

## RESULTS AND DISCUSSION

The PRZM-predicted concentrations and the observed pesticide profiles for the Florida site at 26, 55 and 85 d post-application are presented in Figures 3, 4 and 5. (Metalaxyl residues were not found at the 154-d sampling.) Coefficients of determination ($R^2$) for each of these sampling events are shown in Table 5. Higher values of $R^2$ were observed for the 55- and 85-d samplings, when compared with the 26-d sampling. Integration under the observed and predicted concentration profiles for the 26-d sampling indicated the total area under each profile to be approximately the same, which suggests that mass balance was observed. Differences between the observed and predicted values for the 26-d sampling could be due to a larger hydrodynamic dispersion associated with the first rainfall event than was allowed by the model, or they could be due to sampling error.

Additional movement and dispersion within the soil profile was not appreciable for 55-d (July) and 85-d (August) samplings at the Florida site. (Total precipitation for these months was only 50% of the annual average for Lake City, Florida—therefore, potential recharge and hence pesticide movement would be minimal.) The profile for the 85-d sampling appears to be higher than for the 55-d sampling at the Florida site. The lack of potential recharge and pesticide movement probably allowed the pesticide to remain at the same depth, with only degradative processes acting on the pesticide. The pesticide observed at the lower depths for the 55-d sampling could have degraded to nondetectable levels at the 85-d sampling. (The level of detection was 0.05 ppm.)

The simulated depth of penetration (maximum vertical movement) of metalaxyl was nearly identical to the observed profiles in Figures 4 and 5.

Table 5. Performance testing results for PRZM simulations

| Samplings post-metalaxyl application | Coefficient of determination | |
|---|---|---|
| | Florida | Maryland |
| 1 | 0.33 | 0.75[a] |
| 2 | 0.90 | |
| 3 | 0.95 | |

[a]Overall coefficient of determination for 14 individual observations at a sampling depth of 0–15 cm.

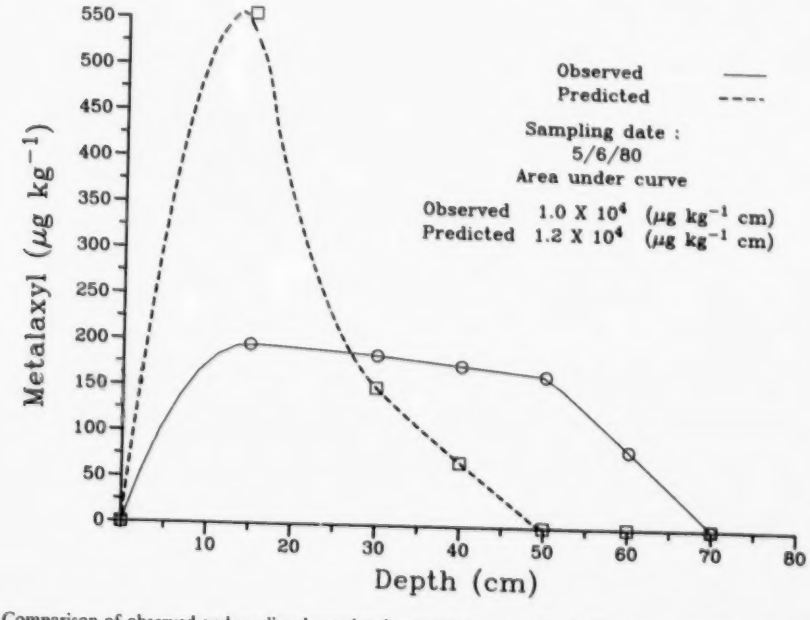


Fig. 3. Comparison of observed and predicted metalaxyl concentration profiles in Florida soil at 26 d post-application.

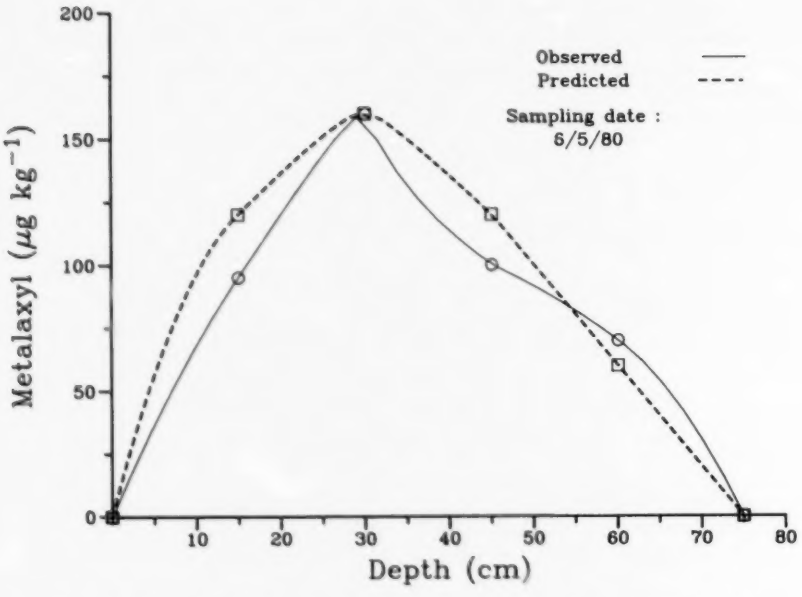


Fig. 4. Comparison of observed and predicted metalaxyl concentration profiles in Florida soil at 55 d post-application.

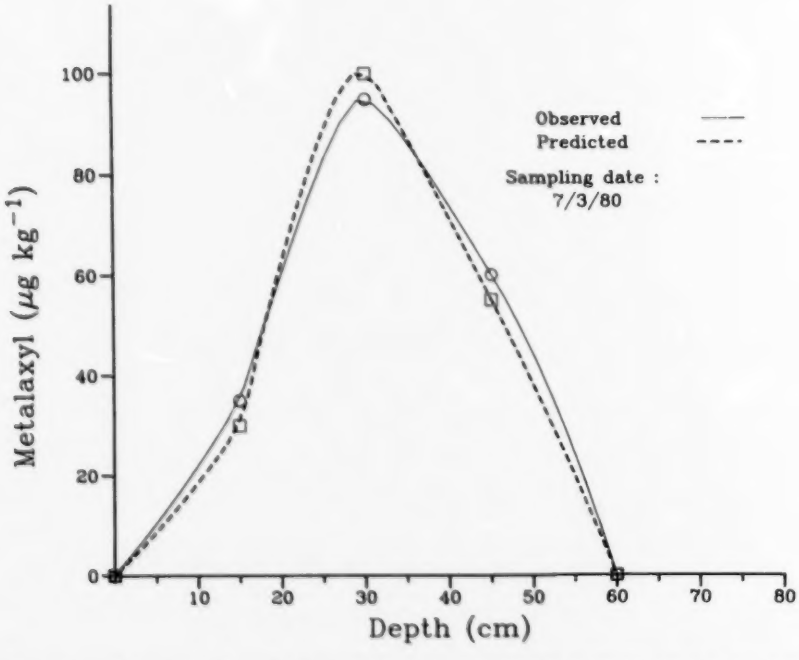


Fig. 5. Comparison of observed and predicted metalaxyl concentration profiles in Florida soil at 85 d post-application.

The depth of maximum penetration may be a useful model performance testing criterion. These results could be used to obtain the mean arrival time to ground water. Arrival times to ground water may have important environmental considerations; however, the estimation of loadings (kg $ha^{-1}$) sufficient to cause health effects may be more important. The first arrival time to ground water could be obtained if the probability density function for water velocity were employed [20].

The PRZM predicted concentration and the observed pesticide profiles for the Maryland site are provided in Figure 6. Metalaxyl did not move deeper than 15.0 cm in the Maryland site. (PRZM predictions also did not indicate movement below 15.0 cm.) The concentration profiles provided in Figure 6 are for the 0 to 15 cm sampling increment only. A coefficient of determination for the 15-, 30-, 44-, 49-, 61-, 76-, 119-, 135-, 152- and 287-d post-application sampling is provided in Table 5. A moderate value of $R^2$ was observed for the sampling events evaluated. The lack of movement in the Maryland site could be attributed to a lack of precipitation for 1980, a higher estimated distribution coefficient (hence more retardation) for metalyaxyl in Maryland, and a lower potential infiltration rate for the Marlton soil. Metalaxyl (mass) remaining in the 0 to 15 cm depth for the 287-d post-application sampling was approximately 10% of both the observed and the predicted values.

Evaluation of PRZM using best estimates for transport and transformation properties of metalaxyl and limited calibration for water balance at the Florida and Maryland sites suggests that the model effectively simulates the important processes operating on the pesticide. PRZM has been successfully tested under field-averaged conditions for citrus and potatoes [7,8] and tobacco. The utility of PRZM for different crops, cropping practices and pesticides has been demonstrated. Additional testing of PRZM is required under conditions of skewed, non-normal distributions to more fully evaluate its predictive capability.

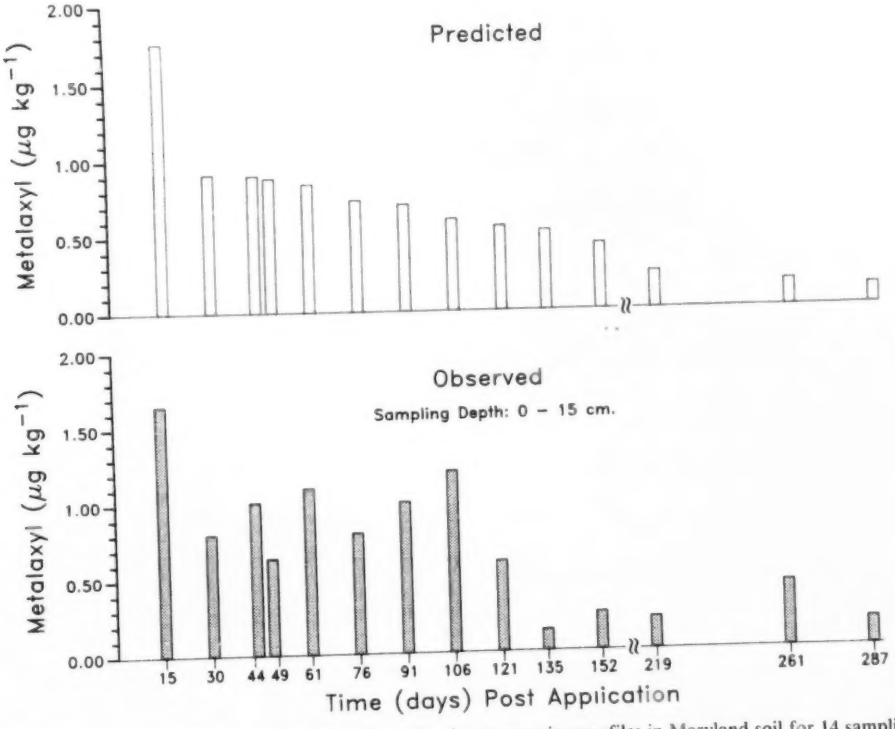


Fig. 6. Comparison of observed and predicted metalaxyl concentration profiles in Maryland soil for 14 samplings post-application.

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